PB90-217399

Mutagenic Atmospheric Aerosol Sources Apportioned by Receptor Modeling

(U.S.) Environmental Protection Agency, Research Triangle Park, NC

1990

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before comple

1. REPORT NO.	2.	3.
EPA/600/J-90/005		PB90-217399

4. TITLE AND SUBTITLE	5. REPORT DATE
MUTAGENIC ATMOSPHERIC AEROSOL SOURCES APPORTIONED BY RECEPTOR MODELING	6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
R.K. Stevens, C.W. Lewis, T.G. Dzubay, R.E. Baumgardner, et al (See title page for other authors)	

9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
U.S. Environmental Protection Agency Atmospheric Sciences Research Laboratory Research Triangle Park, NC 27711 (See Title page for other affiliations)	A101/C/83/02 - 7022 (FY-88) 11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Atmospheric Sciences Research Laboratory - RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

Monitoring Methods for Toxics in the Atmosphere, pp. 187-196, 1990

16. ABSTRACT

During winter 1985 EPA collected air quality samples and data in residential locations in Albuquerque, NM and Raleigh, NC as part of its Integrated Air Cancer Project. Major objectives of study were to evaluate new sampling, analysis and receptor modeling procedures and to apportion fine particle mass and mutagenic activity of the organic fraction of fine particles to appropriate sources. Samples were collected in the two cities for 12-hour periods beginning at 7:00 am and 7:00 pm. This schedule was followed to aid in resolving the organic and inorganic chemical species emitted from residential wood burning (normally a night time activity) and from mobile source emissions. Samples were analyzed for mutagenicity, organic carbon, ^{14}C content and inorganic species. Data from these analyses served as input to a multi-linear regression (MLR model that calculates the relative contribution of extractable particulate organic matter and mutagenic activity observed in these two cities. The ^{14}C results were used to validate the MLR model. Even though the monitoring sites in these cities, were both selected and found to be heavily impacted by residential wood burning (83% & 94% of the organic aerosol mass due to wood burning), a proportionately larger % of the mutagenicity of the fine particles was attributed to mobile sources (25 and 37%). At both sites the mutagenic potency of the organics extracted from fine particles was found to be greater (3-5x's) for mobile sources when compared to wood smoke extractable organics.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
NATIONAL TECHNICAL INFORMATION SERVICE
SPRINGFIELD, VA. 22161

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	12
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE

NTIS is authorized to reproduce and sell this report. Permission for further reproduction must be obtained from the copyright owner.

EPA/600/J-90/005

ASTM Monograph STP-1052, Monitoring Methods for Toxics in the Atmosphere, Walter L. Zielinski, Jr. and William D. Dorko, eds. pp. 187-196, 1990.

R. K. Stevens,[1] C. W. Lewis,[1] T. G. Dzubay,[1] R. E. Baumgardner,[1] R. B. Zweidinger,[1] R. V. Highsmith,[1] L. T. Cupitt,[1] J. Lewtas,[1] L. D. Claxton,[1] L. Currie,[2] G. A. Klouda,[2] and B. Zak[3]

Mutagenic Atmospheric Aerosol Sources Apportioned by Receptor Modeling

REFERENCE: Stevens, R. K., Lewis, C. W., Dzubay, T. G., Baumgardner, R. E., Zweidinger, R. B., Highsmith, R. V., Cupitt, L. T., Lewtas, J., Claxton, L. D., Currie, L., Klouda, G. A., and Zak, B., **"Mutagenic Atmospheric Aerosol Sources Apportioned by Receptor Modeling,"** *Monitoring Methods for Toxics in the Atmosphere, ASTM STP 1052,* W. L. Zielinski, Jr., and W. D. Dorko, Eds., American Society for Testing and Materials, Philadelphia, 1990, pp. 187–196.

ABSTRACT: During the winter of 1985, the U.S. Environmental Protection Agency (EPA) collected air quality data and samples in residential locations in Albuquerque, New Mexico and Raleigh, North Carolina as part of its Integrated Air Cancer Project (IACP). One of the major objectives of these field monitoring studies was to evaluate new sampling, analysis, and receptor modeling procedures and to apportion the fine particle mass and mutagenic activity of the organic fraction of the fine particles to the appropriate sources.

Samples were collected in the two cities for 12-h periods beginning at 7:00 a.m. and 7:00 p.m. This schedule was followed to aid in resolving the organic and inorganic chemical species emitted from residential wood burning (normally a nighttime activity) and from mobile source emissions (normally a daytime activity). Samples were analyzed for mutagenicity, organic carbon, carbon-14 (^{14}C) content, and inorganic characterizations. Data from these analyses were merged into a single data base and served as input to a multilinear regression (MLR) model that calculates the relative contribution of wood burning and motor vehicle sources to the extractable particle organic matter and mutagenic activity observed in these two cities. The ^{14}C measurements were used to validate the MLR model. Even though the monitoring sites in these cities were both selected and found to be heavily impacted by residential wood burning (83 and 94% of the organic aerosol mass due to wood burning), a proportionately larger percentage of the mutagenicity of the fine particles was attributed to mobile sources (25 and 37%). At both sites the mutagenic potency of the organics extracted from the fine particles were found to be greater (three to five times) for mobile sources when compared with wood smoke extractable organics.

KEY WORDS: toxics, receptor modeling, mutagenicity, aerosols, mobile sources, residential wood burning

The mutagenic activity of extractable organic matter (EOM) (EOM is defined as the nonvolatile organic material present in fine particles that are extracted in methylene chloride, using a Soxhlet extraction procedure [*1*]) has been used as a screening tool [*1*] to indicate the potential carcinogenicity of a number of primary emissions from selected combustion sources. There has been a limited attempt to estimate the potential mutagenic impact of combustion emissions on humans through the use of combining conventional predictive

[1] U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.
[2] National Bureau of Standards, Gaithersburg, MD 20899.
[3] Sandia Laboratories, Albuquerque, NM 87185.

models with mutagenicity data on source emissions [2]. The absence of both reliable emission inventories and knowledge on mechanisms of transport and transformation of chemical species responsible for mutagenic activity suggest that a better approach would be to apply receptor modeling [3]. Receptor models are not constrained by these limitations since they have the potential to use the chemical properties of aerosols measured at the source and receptor to calculate the source's contribution to mass or bioactivity of aerosols collected at the receptor.

In order to test the receptor modeling approach, field studies were simultaneously conducted at two geographic locations, Albuquerque, New Mexico and Raleigh, North Carolina, during the 1985 winter heating season. Residential wood burning and mobile source emissions were the only known significant contributors to the fine particle extractable organic matter (EOM) content in both air sheds. The data collected from these two field locations were then used to calculate and validate the relative contribution of mutagenic activity in ambient aerosols from the two sources. The receptor modeling results obtained from the air quality data collected at these two locations are discussed in this paper.

Experimental

Site Descriptions

In 1985, the EPA collected air quality data and samples in two urban locations in Albuquerque and Raleigh. Two primary sampling sites were established at each city. In each city, one of the two sites was located in a residential neighborhood impacted by wood smoke emissions. Numerous single family dwellings with operating fireplaces or wood stoves surrounded these primary sampling sites. The Albuquerque primary woodsmoke site (Zuni Park) is located approximately halfway between downtown and the Eastern Sandia mountain range. Two interstate highways are within 0.7 km of this sampling location. The Raleigh primary woodsmoke site (Quail Hollow Swim Club) is located north of downtown. The site is situated in a topographical depression approximately 1 km from the nearest four-lane highway. Detailed description of the ambient monitoring sites is reported elsewhere [4]. The data reported herein is based on the samples collected at the Albuquerque and Raleigh primary woodsmoke sampling sites.

Sampling Schedule

Aerosol samples were collected in Albuquerque and Raleigh for 12-h periods beginning 7 a.m. and 7 p.m. in order to represent daytime and nighttime conditions. A complete description of the monitoring performed at these sites has been described elsewhere [4]. In Albuquerque, sampling began 12 Jan. and was nearly continuous until 20 Feb. 1985. In Raleigh, samples were collected intermittently from 15 Jan. until 20 March 1985. Most samples in Raleigh were collected when the meteorological forecasts predicted cold periods. Since residential wood burning would be most prevalent during colder periods, higher ambient concentrations of condensable organic species originating from wood stoves would be anticipated in fine particle samples collected during these periods. The Raleigh samples were collected primarily for the development and validation of methods; therefore, we were more interested in the colder sampling periods since one of the objectives of this phase of the project was to collect enough mass of condensable organics to perform multiple mutagenic tests and compare analytical methods that measure a broad range of potentially toxic species present in extractable organic matter.

Sampling and Analysis

Table 1 [*5–13*] is a list of sampling systems and the types of analysis [*5–12*] performed on samples collected in Albuquerque and Raleigh in support of the receptor modeling phase of this study.

After sample collection for carbon-14 (^{14}C) analysis, the quartz filters were shipped in containers, kept at ~5°C, to the EPA Laboratories in Research Triangle Park for storage, analysis, and documentation. The Teflon-coated glass filters were used to obtain the methylene chloride extracts for measuring mutagenic activity and the mass of EOM. A solvent and extraction methods paper describing the selection of an optimum solvent (dichloromethane) and extraction technique are reported elsewhere [*9*]. These filters were stored at −70°C after collection for approximately six months prior to extraction to preserve the sample integrity. The mutagenic activity of the methylene chloride extract was measured by first solvent exchanging the EOM into dimethyl sulfoxide and assaying the organic matter with the *Salmonella typhimurium* plate incorporation assay using strain TA98 both with and without an Aroclor-induced CD-1 rat liver homogenate (+S9). The mutagenic activity was very weak in the absence of S9, and those results are not presented here. The assay was performed according to the methods previously reported [*10*], and it adhered to recently published guidelines [*11*]. Data were analyzed and slope values were generated using the method of Bernstein et al. [*12*]. The uncertainty of these bioassay measurements, based on one half the difference between duplicate determinations, averaged 14%.

Data from the analyzed samples were merged into a single data file for each site for use in the receptor modeling calculations.

^{14}C Analysis

A subset of high-volume fine particle samples collected on quartz filters were selected and sent to the National Bureau of Standards (NBS) for ^{14}C analysis. Details of the analysis procedure have been previously described [*8*]. Due to the relatively high cost of ^{14}C analysis only ten of these samples were analyzed by NBS. These were selected from the 84 samples from Raleigh and Albuquerque that were used in the receptor model calculations described below. Results from the ^{14}C analysis were then compared to source apportionment results obtained statistically with a multiple linear regression (MLR) model. Additional ^{14}C measurements were made on composite samples and selected fractions from extracts of samples collected in this study. Results of these measurements are reported elsewhere [*8*].

Results

Air Quality Data

Average concentrations of the key unique tracer species (Pb for mobile sources and K for wood burning sources [*6*]) used in the receptor model calculations measured in Raleigh and Albuquerque are shown in Table 2. These data indicate that the K concentrations were higher at night than during the day. However, the average Pb concentrations for the paired day-night sampling periods at both sites were similar. Nighttime meteorological inversion conditions reduced the mixing height of the atmosphere which resulted in concentrating the pollutant emitted below the inversion. This probably accounts for the similarities between the day-night Pb concentrations. This concentration pattern suggests mobile sources contributed about the same amount of fine particles during both the day and night, while residential wood burning emissions were mainly at night. Receptor modeling results, dis-

TABLE 1—*Aerosol sampling equipment, collection conditions, and analyses performed in support of receptor modeling.*

Location	Samplers		Flow Rate, L/min	Periods[a]	Filter Media	Measurements		References
	Type	Number				Species	Method	
Zuni Park, Albuquerque								
	Automated sierra (modified)	1	23.0	72	Teflon quartz	mass carbon	gravimetric pyrolysis	5 6
	Beckman dichotomous	1	16.7	72	Teflon	mass elements	gravimetric XRF	5 7
	PM-10 high-volume	2	1000	72	quartz	carbon C-14	pyrolysis accelerator	6 8
	High-volume with 2.5 μm impactor	2	1000	72	Teflon-coated glass	mutagens	Ames test	9–12
	Denuder difference	1	2	72	nylon	nitrate/HNO_3	IC	13
Quail Hollow, Raleigh								
	Sierra dichotomous sampler	1	16.7	57	Teflon	mass	gravimetric XRF	5 7
	High-volume with Sierra impactor inlet (2.5 μm)	2	1000	57	quartz	carbon C-14	pyrolysis accelerator	6 8
	High-volume with Anderson impactor (2.5 μm)	2	1000	57	Teflon-coated glass	mutagens	Ames test	9–12

[a] The duration of all periods was 12 h; 44 of the 72 sampling periods in Albuquerque, NM, and 40 of the 57 periods in Raleigh, NC, were used in the receptor modeling calculations.

TABLE 2—*Average concentration of tracer species used in receptor model calculations for Albuquerque, NM, and Raleigh, NC, studies, micrograms per cubic metre.*

	Albuquerque		Raleigh	
Species	Day	Night	Day	Night
Pb	0.270	0.294	0.082	0.117
K**	0.025	0.105	0.072	0.175

* K* = K corrected for soil contribution.

cussed later, tend to support this conclusion. Previous wintertime air quality data and receptor modeling results from studies performed in Albuquerque and Denver [*14,15*] are similar in many respects to the data and results reported here.

Receptor Modeling Results

The receptor model used to perform the source apportionment of the EOM and mutagenic activity is based on the application of a single tracer multilinear regression model (MLR) developed for a receptor modeling study performed in Denver in 1982 [*14*]. As investigated in previous work [*3,14,15*] in Albuquerque and Denver, we assumed fine particle Pb and K (corrected for soil K) in the winter represented unique tracer species for mobile and residential wood burning emissions. For our MLR calculations, we required a minimum of 30 to 50 observations to minimize the uncertainties in the source apportionment results.

The MLR equation set the dependent variable as either the mass of EOM, in micrograms per cubic metre or mutagenic activity expressed as the number of revertants per cubic metre, R_i, measured for each sampling period, i. The independent variables were K and Pb, and their concentrations are expressed in micrograms per cubic metre. The utility of this approach has been demonstrated, previously, with the Albuquerque data set [*3*] and data collected in Denver, CO [*14*]. For the Albuquerque data set, which consisted of 18 daytime and 26 nighttime observations, the linear regression results for EOM (Eq 1) and mutagenic activity, R_i, (Eq 2) were as follows (observations = 18 days and 26 nights):

$$\mathrm{EOM}_i = (204 \pm 8)\,\mathrm{K}_i + (10.6 \pm 3)\,\mathrm{Pb}_i + (1.0 \pm 0.9) \quad (1)$$

$$r = 0.98$$

$$R_i = (258 \pm 27)\,\mathrm{K}_i + (39 \pm 10)\,\mathrm{Pb}_i + (3 \pm 3) \quad (2)$$

$$r = 0.90$$

Therefore, once these coefficients are determined from the linear regression calculations, the best estimate of the contribution of a source during a particular sampling period is simply the product of the measured tracer concentration and the corresponding regression coefficient. The standard error for the K coefficients and intercepts are relatively small, while the uncertainty in the Pb coefficients are nearly 30%. The larger uncertainty in the Pb coefficient is, in part, associated with the larger number of nighttime samples used in the MLR. However, the multiple correlation coefficient, r, derived from the linear regression model is an indication of the correctness of the model. Additional details related to the MLR model have been discussed previously [*3*].

For the Raleigh data, both the high Br-Pb correlation coefficient (0.98) and Br/Pb ratio (0.27) were similar to those found in Albuquerque, which strongly supports the assumed motor vehicle origin of these elements. However, when we attempted to perform our linear regression analysis on the Raleigh data set, the coefficient for Pb had a large uncertainty. The samples collected in Raleigh and used in the MLR were dominated by wood smoke because the selection of samples for mutagenic analysis used in the receptor model calculations was based on the sample having sufficient mass of EOM available for mutagenic analysis. At the Raleigh site, this meant that the selected samples for MLR calculations were dominated by residential wood burning activities since EOM concentrations were elevated only during cold weather events, a period when wood burning is most prevalent. Therefore, greater than 90% of the EOM for this selected data set was due to wood smoke emissions, making derivation of a reliable Pb coefficient difficult. Because of this problem, we decided to use the coefficient for Pb derived from our Albuquerque study to obtain acceptable MLR results for the Raleigh data set. Since the two studies were contemporary, we felt justified in using the Albuquerque derived Pb coefficient. Below are the linear regression results for Raleigh (observations = 14 day and 26 night):

$$\mathrm{EOM}_i = (112 \pm 4)\,\mathrm{K}_i + (10.6 \pm 3)\,\mathrm{Pb}_i + (0.1 \pm 0.8) \tag{3}$$

$$r = 0.98$$

$$R_i = (88 \pm 7)\,\mathrm{K}_i + (39 \pm 10)\,\mathrm{Pb}_i + (1 \pm 1) \tag{4}$$

$$r = 0.90$$

Again we note the relatively small uncertainty in our K coefficient, small intercepts, and high correlation coefficients, r, for EOM and bioactivity, R_i. These results suggest our use of the Albuquerque Pb coefficient in the MLR analysis of the Raleigh data did not substantially degrade our source apportionment.

The MLR coefficient for K derived from the Raleigh EOM data was significantly smaller than the coefficient derived in Albuquerque. This difference in the K coefficient is consistent with the variations of K emitted by the different types of wood burned in these two cities. In Raleigh, oak or other hardwoods are the major sources of fuel for wood stoves. Potassium emissions from hardwoods are significantly greater than from softwoods (pine) [*6*]. Pinon pine is the principal wood used in Albuquerque; therefore, lower average ambient K concentrations would be expected and were found in Albuquerque than in Raleigh in air sheds impacted by wood smoke.

A jackknife procedure [*15*] was also applied to these data sets to obtain nonparametric estimates of the regression coefficients and their errors. In this procedure, the set of regression results obtained from separately deleting each case is combined to give new estimates of the regression coefficients and their standard errors that are nonparametric, that is, not dependent on the normality assumptions that underlie classical regression. For the 44 observations in Albuquerque and the 40 observations in Raleigh, the jackknife estimates were virtually the same as the values obtained from the standard MLR analysis.

Mutagenic Contribution and Potency of Sources

The ambient measurement-derived potencies of wood smoke and motor vehicle emissions for Albuquerque and Raleigh are derived from the linear regression equations [*1–4*]. They are obtained from the ratio of the mutagenic activity to EOM for each source, and results are summarized in Table 3. The results in Table 3 show the contribution of "other" sources

TABLE 3—*Average ambient concentrations and TA98 + S9 mutagenic potencies attributed to woodsmoke and mobile sources.*

	Woodsmoke	Mobile Sources	Other Sources	Total
Albuquerque results				
EOM, μg/m³	15 ± 1	3 ± 0.9	1 ± 0.9	19
Mutagenicity, revertants/m³	19 ± 2	11 ± 3	3 ± 3.1	32
Potency, revertants/μg EOM	1.3 ± 0.3	3.7 ± 1.5	...	...
Raleigh results[a]				
EOM, μg/m³	16 ± 0.5	1 ± 0.3	0.1 ± 1	17
Mutagenicity, revertants/m³	12 ± 0.9	4 ± 1	1 ± 1	18
Potency, revertants/μg EOM	0.78 ± 0.07	3.7 ± 1.5	...	...

[a] The Pb coefficients were from the Albuquerque results.

to total mutagenicity to be small, indicating that mutagenic activity observed for samples from Albuquerque and Raleigh is almost entirely due to emissions from mobile and residential wood burning sources. In Albuquerque, mobile sources produced 3.7 revertants/μg EOM compared to 1.3 revertant/μg from residential wood burning. In Raleigh, the wood smoke potency was found to be 0.8 revertants/μg EOM, comparable to the Albuquerque result. The motor vehicle potencies are identical, of course, because of the use of the Albuquerque Pb coefficient in Raleigh. The potency values are consistent with the average of mutagenic tests of primary emissions from wood stoves and mobile sources [*1*].

Validation by ^{14}C

The ^{14}C content of ten fine particle samples collected on quartz filters in Albuquerque and Raleigh were analyzed by a new micro analysis procedure developed by NBS [*8*]. The ^{14}C results provide a direct [*16–17*] (nonstatistical) means of measuring the fraction of aerosol carbon that originates from the new or vegetative carbon. Wood burning is assumed to be the source of vegetative carbon since there are no other known sources of fine particle ^{14}C, which is a reasonable assumption for samples collected in the winter in Raleigh and Albuquerque. Table 4 shows a comparison of ^{14}C and MLR results, obtained as follows: The MLR results for Albuquerque were derived using the total carbon measurements (C_t in micrograms per cubic metre) obtained by a combustion [*6*] method along with Pb and K data. The MLR equation for C_t was

$$C_t = (190 \pm 9)\,K_t + (14.1 \pm 3.7)\,Pb_t + (1.4 \pm 1.1) \quad (5)$$

$r = 0.97$ for 44 observations; Pb and K are expressed in micrograms per cubic metre. The percentage woodsmoke values for MLR shown in Table 4 are then simply the ratios of $(190 \pm 9)\,K_t$ to C_t.

For samples collected in Raleigh, the total carbon measurements were not reproducible due to problems associated with the combustion apparatus. However, since the carbon content of the sample was primarily extractable organics (>90%), we chose to use the linear regression coefficients obtained for EOM to calculate the percentage of woodsmoke carbon. Correspondingly, the MLR percentages shown for Raleigh are obtained from the ratios of $(112 \pm 4)\,K_t$ to EOM_t.

The ^{14}C percentages are the NBS-measured fractions of "contemporary" carbon in each sample, divided by a correction factor of 1.10 which relates to the age of the wood [*17*], estimated to be about 50 years in both air sheds. For wood harvested in 1985, the maximum

TABLE 4—*Percent of fine particle carbon from wood burning.*

Location and Sampling Period	^{14}C Results[a,b]	MLR Results
Albuquerque results		
PM[c]	75 ± 6	69 ± 10
PM	66 ± 3	75 ± 11
AM[c]	58 ± 6	52 ± 12
PM	80 ± 5	98 ± 11
AM	61 ± 12	49 ± 13
PM	66 ± 3	61 ± 10
Raleigh results[d]		
PM[e]	84 ± 5	94 ± 9
PM[e]	89 ± 5	94 ± 9
PM	98 ± 5	94 ± 10
PM	73 ± 7	88 ± 9

[a] Total carbon results.
[b] The ^{14}C results have been corrected for the age of the wood.
[c] PM = 7 p.m. to 7 a.m.; AM = 7 a.m. to 7 p.m.
[d] Mass of extractable organics used in MLR calculations for Raleigh, NC samples.
[e] Duplicate ^{14}C analysis.

possible correction is 1.15, which would occur for 31-year-old wood [*18*]. In virtually every case, the ^{14}C and MLR results overlap within the uncertainty limits.

Discussion

A feature of this study that deserves discussion is the determination and source apportionment of the mutagenic potency of the EOM samples from Raleigh and Albuquerque. The samples collected in this study represent the composite contributions of literally thousands of homes and mobile sources, yet the mutagenic potency of the two major source types can be deduced. The mutagenic potency attributed to the mobile sources emissions in Albuquerque, shown in Table 3, were found to be 3.7 revertants/μg. In addition, the mobile source emissions mutagenic potency obtained in this study was three times larger than the value calculated for the wood burning emissions. It is important to note these values, shown in Table 3, are consistent with mutagenic tests of primary emissions from wood stoves and mobile sources performed by a number of investigators [*1,19*]. Thus, from a comparison of these ambient results with the source mutagenic tests, one may conclude that the *net* effect of atmospheric chemical transformations of the primary emissions is small.

Various researchers have examined the effects of atmospheric transformations upon the mutagenicity of the wood smoke particulate emissions [*20–21*]. The effects have been varied, sometimes increasing and sometimes decreasing the mutagenicity of the bulk sample. The agreement between the calculated mutagenic potencies in the air shed and the observed values for the direct source emissions suggests that transformations, if they did occur, had little *net* effect upon mutagenicity of the particulate involved. This result is consistent with smog chamber irradiation experiments [*22*] also conducted as part of the Integrated Air Cancer Project (IACP), which found that simulated atmospheric transformations had *little* or *no* effect upon the mutagenicity of aerosols.

A key feature of this receptor modeling approach was the fact that the study was performed without knowledge of the emission rates or populations or of the mechanisms of transport or transformations of the fine particles. However, data on the K and Pb present in the

primary emissions from residential wood burning and mobile sources, respectively, are a necessary ingredient.

Finally, the new technique for ^{14}C analysis by accelerator mass spectrometry for 50 to 500 μg carbon [*23,24*] was applied to samples collected in Raleigh and Albuquerque. These results serve as an independent direct check on MLR results and provides a means of validating our statistical model.

Acknowledgment

The research described in this article has been funded wholly by the U.S. Environmental Protection Agency. However, it has not been subjected to agency review and therefore does not necessarily reflect the views of the EPA; no official endorsement should be inferred. Furthermore, the mention of commercial products does not constitute endorsement by the U.S. Environmental Protection Agency.

References

[*1*] Lewtas, J. in *Carcinogens and Mutagens in the Environment*, Volume V, H. F. Stich, Ed., CRC Press, Boca Raton, FL, 1988, Chapter 4, pp. 59–74.

[*2*] Lewtas, J., Alfheim, I., and Lofroth, G., "Contribution of Source Emissions to the Mutagenicity of Ambient Urban Air Particles," in review.

[*3*] Lewis, C. W., Baumgardner, R. E., Stevens, R. K., Claxton, L. D., and Lewtas, J., "The Contribution of Woodsmoke and Motor Vehicle Emissions to Ambient Aerosol Mutagenicity," *Environmental Science and Technology*, Vol. 22, 1988, pp. 968–971.

[*4*] Highsmith, R., Baumgardner, R. E., Harris, B., Lumkin, T., Drago, R., Zweidinger, R., and Merrill, R., "The Collection of Wood Combustion Samples in Raleigh, NC and Albuquerque, NM," *Proceedings*, 1987-EPA/APCA Symposium on Measurements of Toxic and Related Air Pollutants, Research Triangle Park, NC, 1987.

[*5*] Courtney, W. J., Shaw, R. W., and Dzubay, T. G., "Precision and Accuracy of a β Gauge for Aerosol Mass Determinations," *Environmental Science and Technology*, Vol. 16, 1982, p. 236.

[*6*] Stevens, R. K., "Sampling and Analysis for Use in Source Apportionment Studies to Determine Impact of Wood Burning on Fine Particle Mass," *Environment International*, Vol. 11, 1985, p. 271.

[*7*] Stevens, R. K., Dzubay, T. G., Russwurm, G. M., and Rickel, D., "Sampling and Analysis of Atmospheric Sulfates and Related Species," *Atmospheric Environment*, Vol. 12(1–3), 1978, p. 55.

[*8*] Klouda, G., Currie, L., Wise, S., Benner, B., and Sheffield, A., "Source Apportionment of Carbonaceous Combustion Products by Micro Radiocarbon Measurements," *Proceedings*, 1987-EPA/APCA Symposium on Measurements of Toxic and Related Air Pollutants, Research Triangle Park, NC, 1987.

[*9*] Williams, R., Pasly, T., Warren, S., Zweidinger, R., Watts, R., Stead, A., and Claxton, L., "Selection of a Suitable Extraction Method for Mutagenic Activity from Woodsmoke-Impacted Air Particles," *International Journal of Environmental and Analytical Chemistry*, in press.

[*10*] Ames, B. N., McCann, J., and Yamasaki, E., "Methods for Detecting Carcinogens and Mutagens with the Salmonella/Mammalian Microsome Mutagenicity Test," *Mutation Research*, Vol. 31, 1975, pp. 347–364.

[*11*] Claxton, L. D., Allen, J., Auletta, A., Mortelmani, K., Nestman, E., and Zeiger, E., "Guide for the *Salmonella typhimurium*/Mammalin Microsome Tests for Bacterial Mutagenicity," *Mutation Research*, Vol. 189, 1987, pp. 83–91.

[*12*] Bernstein, L., Kaldor, J., McCann, J., and Pike, M. C., "An Empirical Approach to the Statistical Analysis of Mutagenesis Data from the Salmonella Test," *Mutation Research*, Vol. 97, 1982, pp. 267–281.

[*13*] Shaw, R. W., Stevens, R. K., Bowermaster, J., Tesch, J., and Tew, E., "Measurements of Atmospheric Nitrate and Nitric Acid; the Denuder Difference Experiment," *Atmospheric Environment*, Vol. 16, 1982, p. 845.

[*14*] Lewis, C. W. and Einfeld, W., "Origins of Carbonaceous Aerosol in Denver and Albuquerque During the Winter," *Environment International*, Vol. 11, 1985, p. 243.

[15] Lewis, C. W., Baumgardner, R. E., Stevens, R. K., and Russwurm, G. M., "Receptor Modeling Study of Denver Winter Haze," *Environmental Science and Technology*, Vol. 20, 1986, p. 1126.

[16] Currie, L. A., Klouda, G. A., Continetti, R. E., Kaplan, I. R., Wong, W. W., Dzubay, T. G., and Stevens, R. K., "On the Origin of Carbonaceous Particles in American Cities: Results of Radiocarbon "Dating" and Chemical Characterization," *Radiocarbon*, Vol. 25, 1983, p. 603.

[17] Currie, L. A., Klouda, G. A., and Gerlach, R. W., "Radiocarbon: Nature's Tracer for Carbonaceous Pollutants," *Residential Solid Fuels. Environmental Impacts and Solutions*, J. A. Cooper and D. Malek, Eds., Oregon Graduate Center Press, Beaverton, OR, 1981, pp. 365–385.

[18] Klouda, G. A., personal communication, National Bureau of Standards, Gaithersburg, MD, December 1987.

[19] Claxton, L. D., "Characterization of Automotive Emissions by Bacterial Mutagenic Bioassay: A Review," *Environmental Mutagenesis*, Vol. 5, 1983, pp. 609–631.

[20] Kamens, R. M., Rives, T. D., Perry, J. M., Bell, D. A., Paylor, R. F., Goodman, R. D., and Claxton, L. D., "Mutagenic Changes in Dilute Wood Smoke as it Ages and Reacts with Ozone and Nitrogen Dioxide: An Outdoor Chamber Study," *Environmental Science and Technology*, Vol. 18, 1984, p. 523.

[21] Bell, D. A. and Kamens, R. M., "Photodegradation of Wood Smoke Mutagens Under Low NO_x Conditions," *Atmospheric Environment*, Vol. 20, 1986, p. 317.

[22] Kleindienst, T. E., Shepson, P. B., Edney, E. O., Claxton, L. D., and Cupitt, L. T., "Wood Smoke: Measurement of Photooxidation Products," *Environmental Science and Technology*, Vol. 20, 1986, p. 493.

[23] Verkouteren, R. M., Klouda, G. A., Currie, L. A., Donahue, D. J., Jull, A. J. P., and Linick, T. W., "Preparation of Microgram Samples on Iron Wool for Radiocarbon Analysis Via Accelerator Mass Spectrometry: A Closed-System Approach," *Nuclear Instruments and Methods*, Vol. B29, 1987, pp. 41–44.

[24] Slota, P. J., Jr., Jull, A. J. P., Linick, T. W., and Toolin, L. T., "Preparation of Small Samples for ^{14}C Accelerator Targets by Catalytic Reduction of CO," *Radiocarbon*, Vol. 29, 1987, p. 303.